Magnetoresistance in Nanostructured Co-Ag Prepared by Mechanical-Alloying

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Abstract **- Fine cobalt and silver powders have been ball-milled to yield a granular solid of composition $Co_{30}Ag_{70}$ in the form of coarse powder. The material is an intimate mixture of grains of fcc Co in a silver matrix. The cobalt is in the single-domain size range, and the samples exhibit coercivity (0.07 T at 296 K) and unusual thermomagnetic effects, including field-induced unidirectional anisotropy. There is a large, isotropic negative magnetoresistance, which exceeds 10 % at 4.2 K.**

I. INTRODUCTION

The observation of very large negative magnetoresistance in nanostructured binary metallic systems has excited much interest in the past two years because of the potential applications. The requirement seems to be that one of the constituents is a 3d-band ferromagnet with markedly different scattering probability for ↑ and ↓ electrons, whereas the other constituent can be a noble metal. Effects are large when the 3d metal is a strong ferromagnet (all 3d↑ states are filled), and the material is inhomogeneous on the scale of the mean free path, which is typically 50 nm for Ag at room temperature.

The first such negative magnetoresistance effects were observed in Fe-Cr multilayers [1], but it was later shown that similar properties could be found in granular magnetic solids produced by sputtering and annealing films of immiscible elements such as Fe-Cu, and Co-Cu [2,3] or by melt spinning [4]. Up to now, observation of the giant magnetoresistance effect has been restricted to thin films and melt-spun ribbons. Here we show that it can also be observed in bulk material, produced by mechanical alloying. We selected the Co-Ag system [5] because the elements are immiscible in all proportions; they form no intermetallic compounds and have no range of solid solution.

II. EXPERIMENTAL METHODS

A 10 g mixture of pure fine powders (Ag - 99.9 %, 2-3 µm ; Co - 99.8 %, < 2 µm) was prepared to give an atomic composition $Co_{30}Ag_{70}$. The mixture was then ball milled with 170 g of 10 mm stainless steel balls in a Fritsch PM5 ball mill for 42 hrs. The result was a free, coarse powder with a grain size of approximately 100 µm. No inhomogeneity on a micron scale was detected in energy-dispersive X-ray analysis in the scanning electron microscope, and the amount of iron impurity was found to be less than 1 %.

The powder was used directly for magnetization measurements in vibrating-sample or SQUID magnetometers (a single grain was used in the SQUID). Electrical measurements were conducted on foils about 3 mm^2 in size obtained by squashing a large single grain, or on small cylinders made by compressing the powder. Four contacts were made with silver paint, and silver leads were used. No heat treatments have been carried out to optimize the observed effects.

III. RESULTS

X-ray diffraction analysis of the samples shows that they are composed of a mixture of silver (a_0 = 0.408 nm) and fcc cobalt (a_0 = 0.358 nm). All diffraction peaks are broad, reflecting both particle-size and strain broadening.

The magnetization curves of $Co_{30}Ag_{70}$ exhibit hysteresis at room temperature (Fig. 1) which is characteristic of non-superparamagnetic particles in the single-domain size range. For hexagonal cobalt, this would mean a size d < 28 nm [6], but the critical size for fcc cobalt may be smaller. The value of the magnetization obtained by extrapolating to zero applied field is $\sigma = 26.1\ JT^{-1}kg^{-1}$, compared with the value of 30.5 $JT^{-1}\ kg^{-1}$ expected from the cobalt content.

Magnetization data at lower temperatures show considerable sensitivity to the thermal history of the sample.

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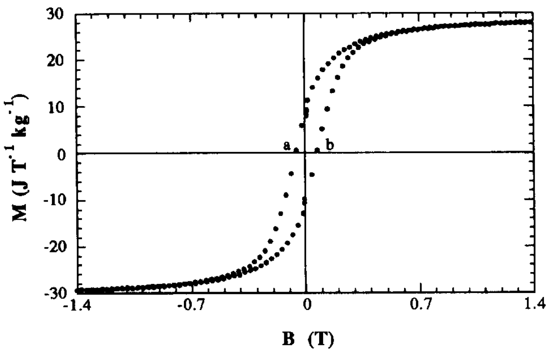

Fig. 1. Hysteresis loop for granular $Co_{30}Ag_{70}$ at 296 K

When field-cooled, the magnetization is easily saturated, but this is not the case for zero-field-cooled samples (Fig. 2). Furthermore, in the SQUID magnetometer measurements where a tiny sample of mass ~10 μg is field-cooled in 8 T, there are giant Barkhausen jumps and evidence of a field-induced anisotropy direction (Fig. 3). Similar giant Barkhausen jumps have been observed in spin glasses [7] and amorphous random-anisotropy magnets [8].

The magnetoresistance data measured at room temperature on a 150 μm foil are shown in Fig. 4. The resistivity of the sample is $3.6 \times 10^{-8}\Omega$m, and this decreases by 3-4 % in 1.4 T, depending on the direction of the field and current. The broader hysteresis when the field is applied perpendicular to the foil can be ascribed to the demagnetizing effect. In the perpendicular geometry, the field is applied perpendicular to the current and to the plane of the foil, whereas in the longitudinal and transverse geometries, the field is applied in the plane of the foil, parallel or perpendicular to the current, respectively. At 4.2 K (Fig. 5) the magnetoresistance in 2.2T is 10 %. The maximum resistivity, equal to the resistivity of the virgin sample, occurs in a field of 0.13 T, which corresponds to the coercive field at 4.2 K (points c and d in figures 2 and 5).

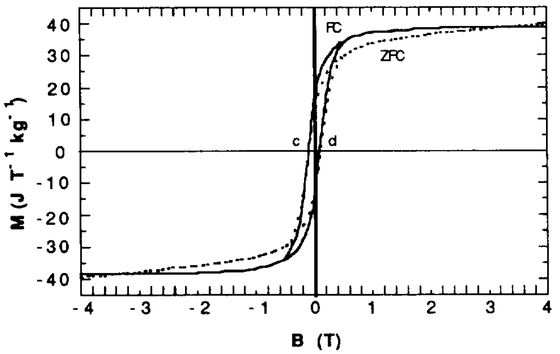

Fig. 2. Hysteresis loop for granular $Co_{30}Ag_{70}$ at 4.2 K, showing the field-cooled (FC) and zero-field-cooled (ZFC) behaviour.

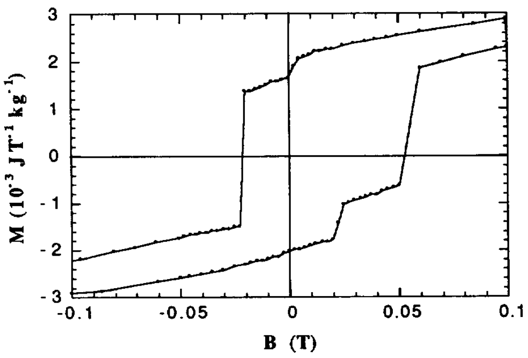

Fig. 3. Hysteresis of a 10 μg sample, exhibiting giant Barkhausen jumps, and a field-induced anisotropy. The sample was cooled from 300 K in 8 T, with the measurement made at 2 K.

IV. DISCUSSION AND CONCLUSIONS

The dependence of magnetization on the thermomagnetic history of these samples suggests that the cobalt particles are coupled by weak magnetic interactions which could be mediated over the spin diffusion length by the Ag 5s conduction electrons. As in a spin glass, it is possible to freeze in a particular magnetic configuration (of the moments

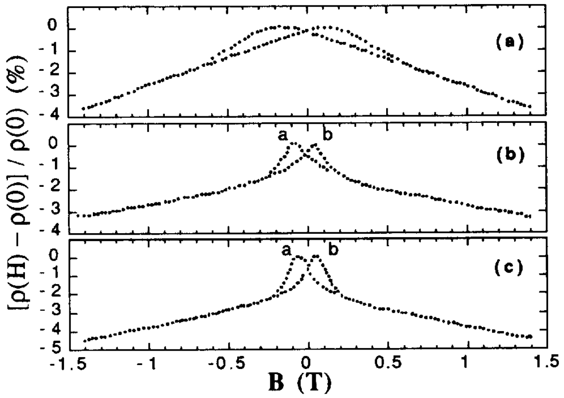

Fig. 4. Magnetoresistance for granular $Co_{30}Ag_{70}$ at 296 K, for different orientations of current and field;
(a) perpendicular, (b) longitudinal and (c) transverse.

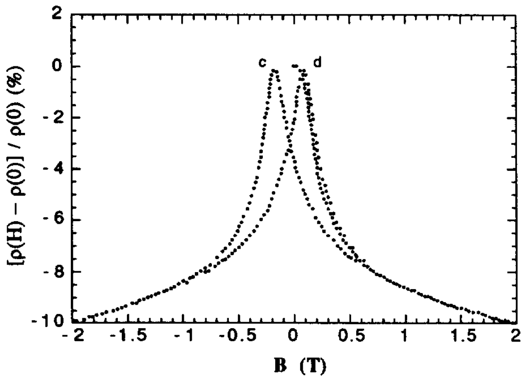

Fig. 5. Transverse Magnetoresistance for granular $Co_{30}Ag_{70}$ at 4.2 K

of the cobalt grains) by field cooling. However, in order to achieve unidirectional anisotropy, it is necessary to have some symmetry-breaking interaction. In spin glasses, the Dzalyoshinsky-Moriya interaction $D.S_i{\times}S_j$ has been suggested [9]. Here we need an interaction between noncollinear spins. A possibility is that the cobalt spins at the Co-Ag interface have an enhanced orbital moment, and therefore adopt a more or less random spin configuration under the action of the local crystal-fields [10].

No attempt has yet been made to optimize the nanostructure by varying the composition or heat treatment to achieve the largest possible magnetoresistance effect. Nevertheless, it is not unreasonable to expect that effects comparable to those achieved in granular films may be achieved in mechanically-alloyed material. The availability of the material in bulk quantities can facilitate certain physical measurements such as neutron diffraction studies, as well as enlarging the scope for exploiting the unusual field-dependent physical properties in practical applications.

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